

# Screened and Unscreened Phases in Sedimenting Suspensions

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A coarse-grained stochastic hydrodynamical description of velocity and concentration fluctuations in steadily sedimenting suspensions is constructed, and analyzed using self-consistent and renormalization group methods. We find that there exists a dynamical, non-equilibrium phase transition from a “unscreened” phase in which we recover the Calfisch-Luke (R.E. Calfisch and J.H.C. Luke, Phys. Fluids **28**, 759 (1985)) divergence of the velocity variance to a “screened” phase where the velocity fluctuations have a finite correlation length growing as  $\phi^{-1/3}$  where  $\phi$  is the particle volume fraction, in agreement with Segrè *et al.* (Phys. Rev. Lett. **79**, 2574 (1997)) and the velocity variance is *independent* of system size. Detailed predictions are made for the correlation function in both phases and at the transition.

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Sedimentation [1] is a rich and complex phenomenon in suspension science and a frontier problem in nonequilibrium statistical mechanics. The average sedimentation speed  $v_{\text{sed}}$  of solute particles drifting down in a solvent is determined by balancing the driving force (gravity) against the dissipative force (viscous drag). Giant non-thermal fluctuations in the velocity and concentration fields in a steadily settling suspension, observed even for non-Brownian systems, have been a puzzle for some years. Calfisch and Luke (CL) [2] showed, for steady sedimentation in a container of smallest linear dimension  $L$ , that the assumption of purely *random* local concentration fluctuations led to velocity fluctuations with a variance  $\langle v^2 \rangle \sim L$ . Most experiments, however, find *no* dependence of  $\langle v^2 \rangle$  on  $L$  [3–5], although Ladd’s simulations [6] and the data of Tory *et al.* [7] appear to be consistent with CL.

In this Letter we propose a resolution of this puzzle by means of a set of coarse-grained, fluctuating nonlinear hydrodynamic equations for the long-wavelength dynamics of concentration and velocity fluctuations in a suspension settling steadily in the  $-z$  direction, at vanishingly small Reynolds number. Our theory is similar in spirit to the Koch-Shaqfeh (KS) [8] “Debye-like” screening approach but differs in several important details and predictions.

The central conclusion of our study is that there are *two* qualitatively distinct nonequilibrium phases for a sedimenting suspension. In the “unscreened” phase  $\langle v^2 \rangle$  diverges as  $L$ , as in CL and, in addition, concentration fluctuations with wavevector  $\mathbf{k} = (\mathbf{k}_\perp, k_z)$  relax at a rate  $\propto k^{1/2}$ . The “screened” phase is characterized by a *correlation length*  $\xi$  similar to that predicted by KS such that  $\langle v^2 \rangle \sim L$  for  $L \ll \xi$  and  $\langle v^2 \rangle \sim \xi$  for  $L \gg \xi$ . Deep in the screened phase we predict  $\xi \sim \phi^{-1/3}$  where  $\phi$  is the particle volume fraction. This is in agreement with the experiments of Segrè *et al.* [5], but not with KS [8]. The relaxation rate in the screened phase is *independent*

of  $k$  for  $k_z = 0$  and  $\mathbf{k}_\perp \rightarrow \mathbf{0}$ . Detailed, experimentally testable expressions for the structure factor and velocity correlations in the screened phase are presented after we outline our calculations. The two phases are separated in our “phase-diagram” (Fig. 1) by a striking *continuous nonequilibrium phase transition* where  $\xi$  diverges at least as rapidly as  $(K - K_c)^{-1/3}$  as a control parameter  $K$  is decreased towards a critical value  $K_c$ .

The hydrodynamic equations we used to arrive at these results are

$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c = [D_{0\perp} \nabla_\perp^2 + D_{0z} \nabla_z^2] c + \nabla \cdot \mathbf{f}(\mathbf{r}, t) \quad (1)$$

and

$$\eta \nabla^2 v_i(\mathbf{r}, t) = m_{Rg} P_{iz} c(\mathbf{r}, t), \quad (2)$$

where  $c(\mathbf{r}, t)$  and  $\mathbf{v}(\mathbf{r}, t)$  are the fluctuations about the mean concentration  $c_0$  and the mean sedimentation velocity  $-v_{\text{sed}} \hat{z}$  respectively. We justify these equations briefly below; for a more detailed discussion we refer the reader to Ref. [9]. Eq. 1 is the anisotropic randomly forced advection-diffusion equation with bare uniaxial diffusivities  $(D_{0z}, D_{0\perp})$  and a random stirring force  $\mathbf{f}(\mathbf{r}, t)$  [10]. The Stokes equation, Eq. 2, which expresses the balance between the driving by gravity and the dissipation by the viscosity  $\eta$ , describes how the concentration fluctuations produce velocity fluctuations. Here  $m_{Rg}$  is the buoyancy-reduced weight of a particle, while the pressure field has been eliminated by imposing incompressibility via the transverse projection operator  $P_{ij} = \delta_{ij} - \nabla_i \nabla_j (\nabla^2)^{-1}$ .

Hydrodynamic equations such as Eqs. 1 and 2 arise from a coarse-graining of the microscopic equations of motion. The latter, for the main case of interest here, *viz.*, non-Brownian suspensions at zero Reynolds number, are the deterministic equations of Stokesian dynamics for  $N$  hydrodynamically coupled particles, and are

known to be chaotic [11]. The noise, or random stirring current  $\mathbf{f}(\mathbf{r}, t)$  and the diffusivities in Eq. 1 represent a phenomenological description of the deterministic chaos at length scales below the coarse-graining length  $\ell$  (which must be large compared to the particle radius  $a$ ). We use these hydrodynamic equations to predict the velocity and concentration fluctuations at length scales large compared to  $\ell$  driven by the random stirring at short distances.

We assume, as is reasonable, that  $\mathbf{f}(\mathbf{r}, t)$  is Gaussian white noise with uniaxial symmetry:

$$\langle f_i(\mathbf{r}, t) f_j(\mathbf{r}', t') \rangle = 2c_0 N_0^{ij} \delta(\mathbf{r} - \mathbf{r}') \delta(t - t') \quad (3)$$

with an anisotropic noise amplitude  $N_0^{ij} = N_{0\perp} \delta_{ij}^\perp + N_{0z} \delta_{ij}^z$ , where  $\delta_{ij}^z$  and  $\delta_{ij}^\perp$  are the projectors along and normal to the  $z$  axis, respectively. Because of the nonequilibrium origin of the noise and diffusion constants, we may not [12] assume that  $N_{0\perp}/N_{0z} = D_{0\perp}/D_{0z}$  as would be true for the Langevin equation of a dilute suspension at thermal equilibrium. Note that no correlations have been fed in via the noise: any that emerge in the long-wavelength properties are a result of the interplay of advection and diffusion.

Let us now consider the nature of the spatio-temporal correlations implied by Eqs. 1 and 2. We will focus on the structure factor for concentration fluctuations

$$S(q) \equiv c_0^{-1} \int d^d r \langle c(\mathbf{0}) c(\mathbf{r}) \rangle e^{-i\mathbf{q} \cdot \mathbf{r}} \quad (4)$$

from which the velocity structure factor can be derived through Eq. 2. If we ignore the advective nonlinearity  $\mathbf{v} \cdot \nabla c$ , then  $S(\mathbf{q})$  can be computed by straightforward Fourier transformation of Eq. 1, resulting in

$$S(\mathbf{q}) = S_0(\mathbf{q}) \equiv \frac{N_{0\perp} q_\perp^2 + N_{0z} q_z^2}{D_{0\perp} q_\perp^2 + D_{0z} q_z^2}. \quad (5)$$

Using Eq. 5 in Eq. 2 we can compute  $\langle v^2 \rangle$  as a function of the system size  $L$  with the result:

$$\langle v^2 \rangle \sim \int_{q>1/L} \frac{S(\mathbf{q})}{q^4} \sim L. \quad (6)$$

In other words, neglecting large-scale advection by the velocity fluctuations leads to the CL [2] result.

To include the effect of the advective nonlinearity we have performed a self-consistent mode coupling calculation [14] on Eqs. 1-3. Our results can be expressed in terms of a *renormalized* relaxation rate

$$R(\mathbf{q}) = D_\perp(\mathbf{q}) q_\perp^2 + D_z(\mathbf{q}) q_z^2 + \Gamma(\mathbf{q}) \quad (7)$$

and a *renormalized* structure factor of the form

$$S(\mathbf{q}) = \frac{N_\perp(\mathbf{q}) q_\perp^2 + N_z(\mathbf{q}) q_z^2}{R(\mathbf{q})}. \quad (8)$$

The quantities  $D_{z,\perp}(\mathbf{q})$  and  $N_{z,\perp}(\mathbf{q})$  represent renormalized diffusivities and noise amplitudes [13]. But, most importantly, the advective nonlinearity to lowest-order perturbation theory leads to an additional term in the renormalization of the relaxation rate which is of the form  $\Gamma(\mathbf{q}) = \gamma(\mathbf{q}) q_\perp^2 / q^2$ . Starting from the stochastic hydrodynamic equations, Eqs. 1-3, it turns out that the amplitude of this singular contribution becomes a constant,  $\lim_{q \rightarrow 0} \gamma(\mathbf{q}) \propto I(\beta_N, \beta_D)$ , which depends on the anisotropy ratios of the noise and diffusivity coefficients

$$\beta_N = \frac{N_\perp}{N_z}, \quad \text{and} \quad \beta_D = \frac{D_\perp}{D_z}. \quad (9)$$

In particular  $I(\beta_N, \beta_D)$  is proportional to  $\beta_N - \beta_D$ . and consequently may change sign upon varying the noise and diffusivity ratios. For  $I(\beta_N, \beta_D) < 0$  this would lead to exponentially growing concentration fluctuations in the limit of long wavelength. Here we do not pursue this intriguing possibility further but instead restrict our attention to  $I(\beta_N, \beta_D) \geq 0$ , for which the model can either be treated within dynamic renormalization group theory or using self-consistency methods.

We start our discussion at the borderline of stability,  $\beta_N = \beta_D$ . For these parameter values it can be shown that the fluctuating hydrodynamic equations describe a dynamics which obeys detailed balance [15]: the advective nonlinearity does not affect the equal-time correlations, and  $S(\mathbf{q})$  in particular is just the constant  $N_\perp / D_\perp$ . There are singularities in  $N_{\perp,z}$  and  $D_{\perp,z}$  which we discuss later.

For  $\beta_N \geq \beta_D$ , detailed balance is violated and a singular diffusion term  $\Gamma(\mathbf{q})$  is generated within perturbation theory. In order to analyze the dynamics in this regime we use one-loop self-consistent theory (mode coupling theory) and arrive at the expression

$$\Gamma(\mathbf{q}) = c_0 \left( \frac{m_R g}{\eta} \right)^2 \int_k \frac{q_i P_{iz}(\mathbf{k}) k_j P_{jz}(\mathbf{q})}{k^2 q^2} \times \frac{[S(\mathbf{q} - \mathbf{k}) - S(\mathbf{k})]}{R(\mathbf{k}) + R(\mathbf{q} - \mathbf{k})} \quad (10)$$

with  $R(\mathbf{q})$  given by (7), and similar self-consistent integral equations for  $D_\perp(\mathbf{q})$ ,  $D_z(\mathbf{q})$ ,  $N_\perp(\mathbf{q})$ , and  $N_z(\mathbf{q})$ . We find that there are two types of iteratively stable solutions to these coupled self-consistent equations: those with  $\gamma(q \rightarrow 0) > 0$ , which we obtain below the solid line in the phase diagram spanned by the two anisotropy ratios (“screened” phase in Fig. 1), and those with  $\gamma(q \rightarrow 0) = 0$ , which arises for values of the anisotropy parameters that lie above the solid line and below the dashed line of the same figure, i.e., in the “unscreened” phase. Note that within the self-consistent theory the line in the phase diagram where  $\gamma(q = 0)$  changes sign (solid line) has shifted with respect to the result of the one-loop perturbation theory discussed above (dashed line).

*Screened Phase:* In the screened phase,  $\Gamma(\mathbf{q})$  is of the form  $\gamma q_{\perp}^2/q^2$  in the small  $q$  limit, with  $\gamma$  a finite constant. This implies that the structure factor at small wavenumber becomes

$$S(\mathbf{q}) \simeq \frac{N_{\perp} q_{\perp}^2 + N_z q_z^2}{D_{\perp} q_{\perp}^2 + D_z q_z^2 + \gamma q_{\perp}^2/q^2} \quad (11)$$

with  $N_{\perp,z}$  and  $D_{\perp,z}$  constants. From Eq. 11 we can define a correlation length  $\xi \equiv (D_{\perp}/\gamma)^{1/2}$  such for  $q_{\perp} \gg 1/\xi$  the structure factor is not significantly affected by advection. On the other hand, for  $q_{\perp} \ll 1/\xi$  the in-plane structure factor reads  $S(\mathbf{q}_{\perp}, q_z = 0) \simeq (N_{\perp}/\gamma) q_{\perp}^2$ , while  $S(\mathbf{q}_{\perp} = 0, q_z) \simeq (N_z/D_z)$ . Physically, this means that at long wavelength advection strongly suppresses in-plane concentration fluctuations.

Using Eq. 11 in conjunction with Eq. 2, one finds that for length scales  $L$  less than  $\xi$ ,  $\langle v^2 \rangle \propto L$ , consistent with CL, while for  $L$  large compared to  $\xi$ ,  $\langle v^2 \rangle \propto \xi$ . Velocity fluctuations on length scales small compared to  $\xi$  are thus highly correlated while they become uncorrelated at larger length scales.

Deep inside the screened phase, i.e., for large  $\gamma$ , the renormalization of the diffusion and noise parameters is negligible and we can explicitly compute  $\gamma$ , and thus  $\xi$ , by inserting Eq. 8 in Eq. 10 using the bare values for the  $N$ 's and  $D$ 's. We find

$$\xi = 8 \left( \frac{m_R g}{\eta D} \right)^{-2/3} c_0^{-1/3} \left( 1 - \frac{2}{\beta_N} \right)^{-1/3}, \quad (12)$$

where for simplicity we have set  $D_{0\perp} = D_{0z} = D$ . According to Eq. 12, the correlation length increases as we decrease the  $\beta_N$  parameter (which could be done by increasing the *thermal* noise amplitude) and diverges at  $\beta_N = 2$ . Strictly speaking, as  $\beta_N \rightarrow 2$ , the diffusivity corrections are no longer negligible, and the actual divergence of  $\xi$  is probably stronger than (12), and occurs at a larger value of  $\beta_N$ . An explicit analytical (but lengthy) result for the correlation length  $\xi$  can also be obtained throughout the screened phase as a function of both anisotropy parameters [9] and the phase boundary can also be computed. The phase boundary resulting from this result is shown in Fig. 1 as the solid line separating the screened from the unscreened phases. The dashed line in the figure corresponds to the set of parameter values where the hydrodynamic equations correspond to a Langevin dynamics in thermal equilibrium.

*Unscreened Phase:* As already noted above, the hydrodynamic equations obey detailed balance [15] along the line  $\beta_N = \beta_D$  in the phase diagram. As a consequence the ratio of noise to diffusivity can be identified as a direction-independent “noise-temperature”. Furthermore, the structure factor  $S(\mathbf{q})$  becomes a constant  $D_{\perp}/N_{\perp}$  and we recover the CL result. In conjunction with an exponent identity resulting from Galilean invariance this is enough to determine the dynamic exponent

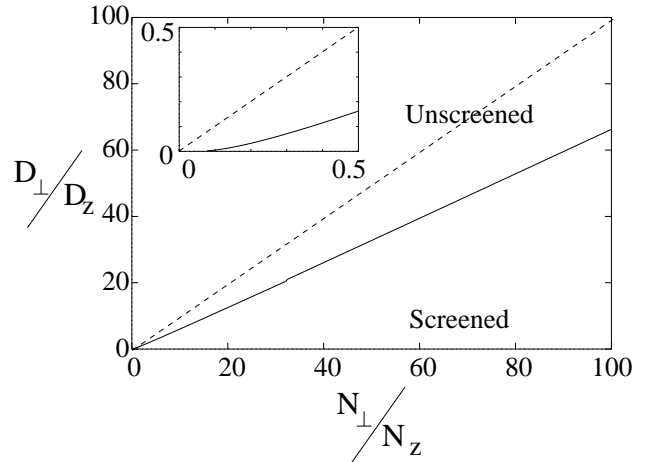


FIG. 1. Dynamical phase diagram for sedimentation. Below the solid line in the parameter space spanned by the anisotropy factors for the noise and diffusivities velocity and concentration fluctuations have a finite screening length in the limit vanishing wavevector. This region is called “screened” above. In the upper region called “unscreened” the screening length becomes infinite. The dashed line represents the set of values for the anisotropy factors where the hydrodynamics obeys detailed balance. The inset shows the behavior of the phase boundary in the limit of large noise and diffusivity in the vertical direction as compared to the horizontal plane.

exactly,  $z = d/2 - 1$ . This implies that the diffusivities and noise amplitudes scale as  $q^{-\epsilon/2} = q^{-3/2}$  for long wavelength. Even though there are now singular corrections to  $D_{z,\perp}(\mathbf{q})$  and  $N_{z,\perp}(\mathbf{q})$ , the anomalous  $\Gamma(\mathbf{q})$  term is zero. For parameter values in the regime between the dashed line (detailed balance line) and the solid line, which marks the location of the nonequilibrium phase transition, renormalization group methods may be used to determine the renormalization of the noise and diffusivity amplitudes. In view of the results from the above self-consistency calculation ( $\gamma = 0$  in the unscreened phase) and the exact results at the detailed balance line it is quite likely that the resulting renormalization group flow will tend towards a fixed point which obeys detailed balance. We leave the details of such an investigation for a future publication [9].

The analysis of our hydrodynamic equations thus confirms that screening can suppress the CL divergence of  $\langle v^2 \rangle$  with  $L$ , as argued by KS, while it allows for a second, unscreened phase. This result may help explain the conflicting results on  $\langle v^2 \rangle$  obtained by different workers [3–7]. The self-consistent structure factor, Eq. 5 we obtained differs significantly from the one proposed by KS. Experimental test will thus be of considerable importance. Measurements of  $S(\mathbf{q})$ , for example by PIV [16] (Particle Imaging Velocimetry), would constitute the most direct test of the theory since our prediction that  $S(\mathbf{q}_{\perp}, q_z = 0) \propto q_{\perp}^2$  does not hold in the KS description. Detailed measurements of  $S(\mathbf{q})$  for sedimenting solutions are not yet available. However, Segrè *et al.* [5] do report

that the size-dependence of the amplitude  $\langle v^2 \rangle$  of the velocity fluctuations depends on a characteristic length scale  $\xi_S$  such that  $\langle v^2 \rangle \propto \xi_S$  for length scales  $L \gg \xi_S$  while for  $L \ll \xi_S$ ,  $\langle v^2 \rangle$  grows with  $L$ . They report that  $\xi_S \sim a\phi^{-1/3}$  with  $\phi$  the particle volume fraction.

Our correlation length  $\xi$ , in Eq. 12, has the same physical interpretation as  $\xi_S$ . Deep in the screened phase, i.e., for  $I(\beta_N, \beta_D) \gg 0$ ,  $\xi$  can be written as:

$$\xi(\phi) \sim (m_R g / \eta D)^{-2/3} a \phi^{-1/3} I(\beta_N, \beta_D)^{-1/3} \quad (13)$$

On scaling grounds, we expect that  $D \propto \delta v_{\text{RMS}} \xi$  with  $\delta v_{\text{RMS}}$  the root mean square of the velocity field fluctuations. Experimentally,  $\delta v_{\text{RMS}} \xi$  is found to be independent of volume fraction  $\phi$ . In that case, Eq. 13 reproduces the experimentally observed volume-fraction dependence, in contrast to KS [8]. It should be noted that this volume fraction dependence of the correlation length implies that there is a fixed number of colloids within a correlation volume independent of volume fraction.

The observation of a transition from the screened to the unscreened phase would obviously be the most conclusive evidence supporting our theory, in particular if the transition were accompanied by a divergence of the velocity fluctuation correlation length. Even in the absence of such direct evidence, the observation of screened behavior combined with our theory requires that the anisotropies in the noise and diffusivity lie in the lower region of our dynamical phase diagram, Fig. 1. A complete test of our theory thus requires measurement of the  $N$  and  $D$  parameters. These could be obtained from the measurement of the steady-state static structure factor  $S(\mathbf{q})$ , e.g. by particle imaging or light scattering experiments both along the  $z$  direction and in the  $x-y$  plane, coupled with tracer diffusion measurements.

Finally, it would be interesting to vary the effective noise and diffusion constants in a controlled manner in an experiment. While there is, as yet, no method to calculate these constants directly from a microscopic theory it is reasonable to expect that by decreasing the Peclet number (i.e., increasing the role of *isotropic* thermal diffusion) one could drive the sedimenting system into the unscreened phase. Thus by repeating the experiments of Segrè *et al.* [5] with colloids that are more nearly density matched to the solvent one could test our prediction of a transition to an unscreened phase.

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  - [13] Including frequency-dependence will not alter the critical exponents and the structure of the scaling variable; it may, however, affect the functional form of the full relaxation rates.
  - [14] The “Galilean” invariance of Eqs. 1 and 2 under  $\mathbf{v}(\mathbf{r}, t) \rightarrow \mathbf{v}(\mathbf{r}, t) + \mathbf{U}$ ,  $\mathbf{r} \rightarrow \mathbf{r} - \mathbf{U}t$  guarantees that the nonlinear coupling will not renormalize. This means we need to worry only about the corrections to the noise strength and relaxation rate; A. Levine, S. Ramaswamy, E. Frey, and R. Bruinsma (unpublished); see also D. Forster, D.R. Nelson, and M.J. Stephen, *Phys. Rev. A* **16**, 732 (1977).
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